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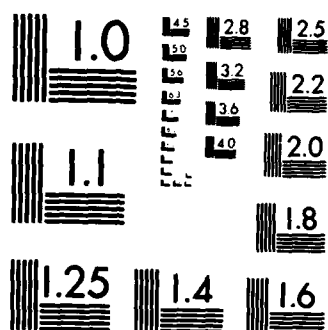UNCLASSIFIED DAAG29-83-C-0005   F/G 4/2   NL

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REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
1. REPORT NUMBER ARO 19893.4-GS	2. GOVT ACCESSION NO. N/A	3. RECIPIENT'S CATALOG NUMBER N/A
4. TITLE *(and Subtitle)* Time and Space Variability of Turbulent Concentration Fluctuations		5. TYPE OF REPORT & PERIOD COVERED Final Report 3 Jan.1983 through 2 Jan 85
		6. PERFORMING ORG. REPORT NUMBER
7. AUTHOR(s) Steven R. Hanna		8. CONTRACT OR GRANT NUMBER(s) DAAG29-83-C-0005
9. PERFORMING ORGANIZATION NAME AND ADDRESS Environmental Research & Technology, Inc. 696 Virginia Rd. Concord, MA 01742		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS N/A
11. CONTROLLING OFFICE NAME AND ADDRESS J. S. Army Research Office Post Office Box 12211 Research Triangle Park, NC 27709		12. REPORT DATE 31 December 1984
		13. NUMBER OF PAGES 7 + 22 page Appendix
MONITORING AGENCY NAME & ADDRESS(If different from Controlling Office)		15. SECURITY CLASS. (of this report) Unclassified
		15a. DECLASSIFICATION/DOWNGRADING SCHEDULE

DISTRIBUTION STATEMENT (of this Report)

Approved for public release; distribution unlimited.

FEB 11 1985

DISTRIBUTION STATEMENT (of the abstract entered in Block 20, If different from Report)

NA

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18. SUPPLEMENTARY NOTES

The view, opinions, and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy, or decision, unless so designated by other documentation.

19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

TURBULENCE, DIFFUSION, CONCENTRATION FLUCTUATIONS, SMOKE WEEK EXPERIMENTS.

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Models for concentration fluctuations in atmospheric smoke plumes are reviewed and a simplified model developed. The new model is shown to agree well with Smoke Week III data and fluid modeling data.

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TIME AND SPACE VARIABILITY OF

TURBULENT CONCENTRATION FLUCTUATIONS

FINAL REPORT

by

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DECEMBER 31, 1984

U.S. ARMY RESEARCH OFFICE

CONTRACT NUMBER DAAG29-83-C-0005

ENVIRONMENTAL RESEARCH AND TECHNOLOGY, INC.

696 VIRGINIA ROAD, CONCORD, MA 01742

REPORT NUMBER PB-811

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1. STATEMENT OF PROBLEM STUDIED

Observed and modeled concentration fluctuations in smoke plumes in the atmosphere are quite large, with the standard deviation σ_c at least as large as the mean $\overline{C}$. There is interest in concentration fluctuations because of their importance in assessing environmental and toxicological effects, in evaluating models for predicting mean concentrations, and in determining the response of remote sensors pointed towards smoke plumes. During the past ten years there has been a large increase in research on this subject because of the concerns listed above, because of advances in the development of instruments than can measure short-term concentration fluctuations, and became of increases in speed and storage capabilities of computers that some researchers are using to run models of concentration fluctuations.

The purpose of the research performed under this contract is to review existing models and data sets on concentration fluctuations, and use this information to develop and test a simplified model. Emphasis is on the use of U.S. Army data to evaluate the model. As a result of the model evaluation, research required for model improvement is recommended and steps taken to begin some of this new research.

2. SUMMARY OF THE MOST IMPORTANT RESULTS

This research project can be divided into three tasks:

- Review of existing models and data,
- Development of simple model and evaluation with U.S. Army data.
- Development of strand model.

The first two tasks were completed during the first 20 months of the project and most of the results are reported in the publications that are listed in Section 3. Abstracts of the journal articles are given here, and the reader is referred to the original articles for details.

The last task has been underway since September, 1984, and is not completed. It is briefly reviewed below and a more detailed progress report is given as Appendix A.

2.1 Review of Existing Models and Data

CONCENTRATION FLUCTUATIONS IN A SMOKE PLUME
Steven R. Hanna

Abstract – Previous research results are reviewed and used to derive a new set of analytical formulas for predicting concentration fluctuations in smoke plumes. The meandering plume approach and the internal plume approach are compared. Some simple models are tested with field and laboratory data sets, showing that several aspects of the data (e.g. the concentration fluctuations on the plume axis) are reasonably well simulated. However there is much room for improvement, since the models have some fundamental disagreements and must more testing with data should take place. In particular, a good knowledge of the Lagrangian time scale is essential for predicting concentration fluctuations.

2.2 Development of Simple Model and Evaluation with U.S. Army Data

THE EXPONENTIAL PROBABILITY DENSITY FUNCTION AND CONCENTRATION FLUCTUATIONS IN SMOKE PLUMES
Steven R. Hanna

Abstract – Observations of 1-s average concentration fluctuations during two trials of a U.S. Army diffusion experiment are presented and

compared with model predictions based on an exponential probability
density function (pdf). The source is near the surface and
concentration monitors are on lines about 30 to 100m downwind of the
source. The observed ratio of the standard deviation to the mean of
the concentration fluctuations is about 1.3 on the mean plume axis and
4 to 5 on the mean plume edges. Plume intermittency (fraction of
non-zero readings) is about 50% on the mean plume axis and 10% on the
mean plume edges. A meandering plume model is combined with an
exponential pdf assumption to produce predictions of the intermittency
and the standard deviation of the concentration fluctuations that are
within 20% of the observations.

In addition to the above paper that was published in Boundary
Layer Meteorology, three other papers have been prepared in which the
simple model is compared with observations. two of these can be found
in conference proceedings (CRDC Conference, AMS Conference), and the
third is to be published soon in Atmospheric Environment. The first
two present additional statistics from the U.S. Army Smoke Week III
experiment, including observed probability density functions and
moments of the distributions of instantaneous concentration, plume
width, plume centroid, and cross-wind integrated concentration. The
latter paper compares the new model with observations of ground level
concentrations resulting from tracer releases from a tall stack in
Deardorff and Willis' convective tank.

2.3 Development of Strand Model

The strand model is a new concept that is under development and
has not yet been published in a journal or report. A preliminary
description of the model is in Appendix A. The purpose of this
research is to explicitly account for concentration fluctuations at
very small scales. This model assumes that the source emits strands
of polluted material, which diffuse due to molecular motions and split
when their diameters double. If polluted strands are close enough
together, they may entrain each other when they split. The whole
packet of strands also diffuses at a rate comparable to that of

3

an instantaneous Gaussian puff. When combined with a model for the
meander of the instantaneous plume, the model gives predictions in
agreement with wind tunnel data of Fackrell and Robins. The model
produces a probability density function of the concentration
fluctuations at a point, and can account for the effects of source and
sampler size and sampling and averaging time.

3. LIST OF PUBLICATIONS

Hanna, S.R., 1984. Concentration Fluctuations in a Smoke Plume.
    Atmos. Environ., 18, 1091-1106.

Hanna, S.R., 1984. The Exponential Probability Density Function and
    Concentration Fluctuations in Smoke Plumes. Bound. Lay.
    Meteorol., 29, 361-375.

Hanna, S.R., 1984. Observed and Modeled Concentration Fluctuations in
    a Small Smoke Plume. Proceedings of Fourth Joint Conference on
    Applications of Air Pollution Meteorology. Am. Meteorol. Soc.,
    128-131.

Hanna, S.R., 1984. Comments on Deardorff and Willis' (1984) Paper
    Entitled "Ground-Level Concentration Fluctuations from a Buoyant
    and a Non-Buoyant Source within a Laboratory Convectively-Mixed
    Layer." to be published in Atmos. Environ.

Hanna, S.R. and J. Pleim, 1984. Characteristics of Observed
    Concentration Fluctuations During Smoke Week III. Proceedings of
    1984 CRDC Scientific Conference on Obscuration and Aerosol
    Research, Aberdeen Proving Ground, MD.

4. LIST OF MEETINGS ATTENDED

Feb. 20-21, 1983, White Sands Missile Range.

Concentration fluctuation data sets were discussed with W. Ohmstede, R. Sutherland, and others at the Atmospheric Science Laboratory. A seminar on models of concentration fluctuations was given. Plans were made for a possible smoke experiment to take place this fall.

June 15-17, 1983, ARO Quail Roost Workshop on Aerosol Dispersion in the Atmospheric Surface Layer.

At this workshop current U.S. Army research projects were discussed and recommendations made for future research priorities.

November 17, 1983, Aberdeen Proving Ground, MD.

A paper entitled "Concentration fluctuations in smoke plumes from near surface releases" was presented at the 1983 Scientific Conference on Chemical Defense Research. This research project was discussed with D. Sloop and R. Saucier of CSL.

December 5 & 6, 1983 and February 3, 1984, Boulder, CO.

Dr. Hanna attended a meeting of the Large Eddy Simulation Working Group, sponsored by ARO. Research plans for this project were discussed with W. Bach of ARO and W. Ohmstede of ASL.

June 25, 1984, Aberdeen Proving Ground, MD.

Dr. Hanna attended the 1984 CRDC Scientific Conference on Obscuration and Aerosol Research at Aberdeen, Proving Ground, MD, where he presented a paper entitled "Characteristics of Observed Concentration Fluctuations during Smoke Week III.

October 19, 1984, Portland, OR

> Dr. Hanna presented a paper entitled "Observed and Modeled
> Concentration Fluctuations in a Small Smoke Plume" at the
> AMS/APCA Fourth Joint Conference on Applications of Air Pollution
> Meteorology.

October 23-25, 1984, Kiawah Island, SC.

> Dr. Hanna attended the DOE/AMS Workshop on Model Evaluation in
> Kiawah Island, SC, where he presented a talk on the results of
> this project as they affect model evaluation, and discussed
> progress with Dr. Bach of ARO.

November 7, 1984, Raleigh, NC

> Dr. Hanna visited Dr. Bach and Dr. Flood at ARO in RTP, NC.

December 5, 1984, White Sands Missile Range, NM.

> Dr. Hanna visited ASL at White Sands Missile Range, NM, where he
> presented a seminar on his research and discussed this project
> with R. Meyers and other ASL scientists.

5. LIST OF PARTICIPATING SCIENTIFIC PERSONNEL

> Dr. Steven R. Hanna
> Dr. Robert J. Yamartino
> Mr. Jonathan Pleim

No degrees were awarded.

APPENDIX A

A STRAND THEORY OF CONCENTRATION FLUCTUATIONS

by

R.J. Yamartino

and

S.R. Hanna

STATUS REPORT

1. Introduction

The notion that pollutants may be emitted, dispersed and
described mathematically as though the material were confined to small
packets of size λ has been described by Csanady (1973), Chatwin and
Sullivan (1979), Venkatram (1983), and recently by Chatwin (1984).
The prime motivation for such a formulation is that higher moments of
the concentration distribution, including the variance, can be readily
computed since the problem reduces to that of "counting" statistics.

The size of the packets, λ, is generally chosen to correspond
to the conduction length scale where molecular diffusion is the only
operative dilution mechanism; however, most treatments then either
ignore the effects of molecular diffusion or assume that packet growth
by molecular diffusion results only in the entrainment of clean air.

In this paper we assume that pollutants emerge from a source in
long thin strands, rather than small three-dimensional packets, and
that these strands subsequently grow by diffusive processes until a
time t_d, whereupon they split into two strands that then repeat the
above mentioned process. Consideration of strand entrainment of both
clean and polluted air leads to a differential equation for the time
development of the spatial density of polluted strands. The solution
of the resulting first-order, non-linear, differential equation
combined with counting statistics then permits a determination of
ensemble average concentration statistics as would be measured by a
finite aperture detector possessing a time response that is extremely
rapid compared with all relevant time scales.

Model predictions for plume intermittency are then compared with
the wind tunnel observations of Fackrell and Robins (1982). Further
assumptions about the statistical nature of the strands then permits
total and conditional (i.e., $C>0$) concentration statistics to be
evaluated. Additions to the theory to include plume meander, time
averaging, and finite response detectors are discussed.

2. Strand Theory

2.1 Development of the Governing Equations

Rather than postulating a non-linear differential equation for
the concentration variance, as is done by Lewellen and Sykes (1983),
we begin with the simple notion of pollutants confined to thin strands
of diameter λ that are dispensed by turbulent eddies and also grow
in size until shear forces tear the strand in two. Such an approach
leads naturally to the computation of higher moments of the observed
concentration distribution including the variance. The actual size of
these strands will correspond to the conduction length scale, which
Chatwin and Sullivan (1979) estimate to be of order 10^{-3} meter;
however, specific choice of a numerical value for λ is not a
prerequisite for development of the formalism.

Consider a source of diameter d_s emitting at rate Q (mass/time)
into a flow field of velocity u. Assuming that the emissions are well
mixed across the source aperture, one might imagine (Figure 1) that
the pollutant emerges from the source in thin strands of diameter λ
and containing a concentration C_o. In such a case the number of
strands, N_s, emerging would be

$$N_s = (d_s/\lambda)^2 \tag{1}$$

and the concentration in the strands would be

$$C_o = Q/(\frac{\pi}{4} d_s^2 u) \tag{2a}$$

or

$$C_o = Q/[2\pi \sigma_y(0) \sigma_z(0) u] \tag{2b}$$

in the more familiar notation of plume modeling. Instantaneous
concentrations $C(x,y,z)$ downwind could then be expressed as
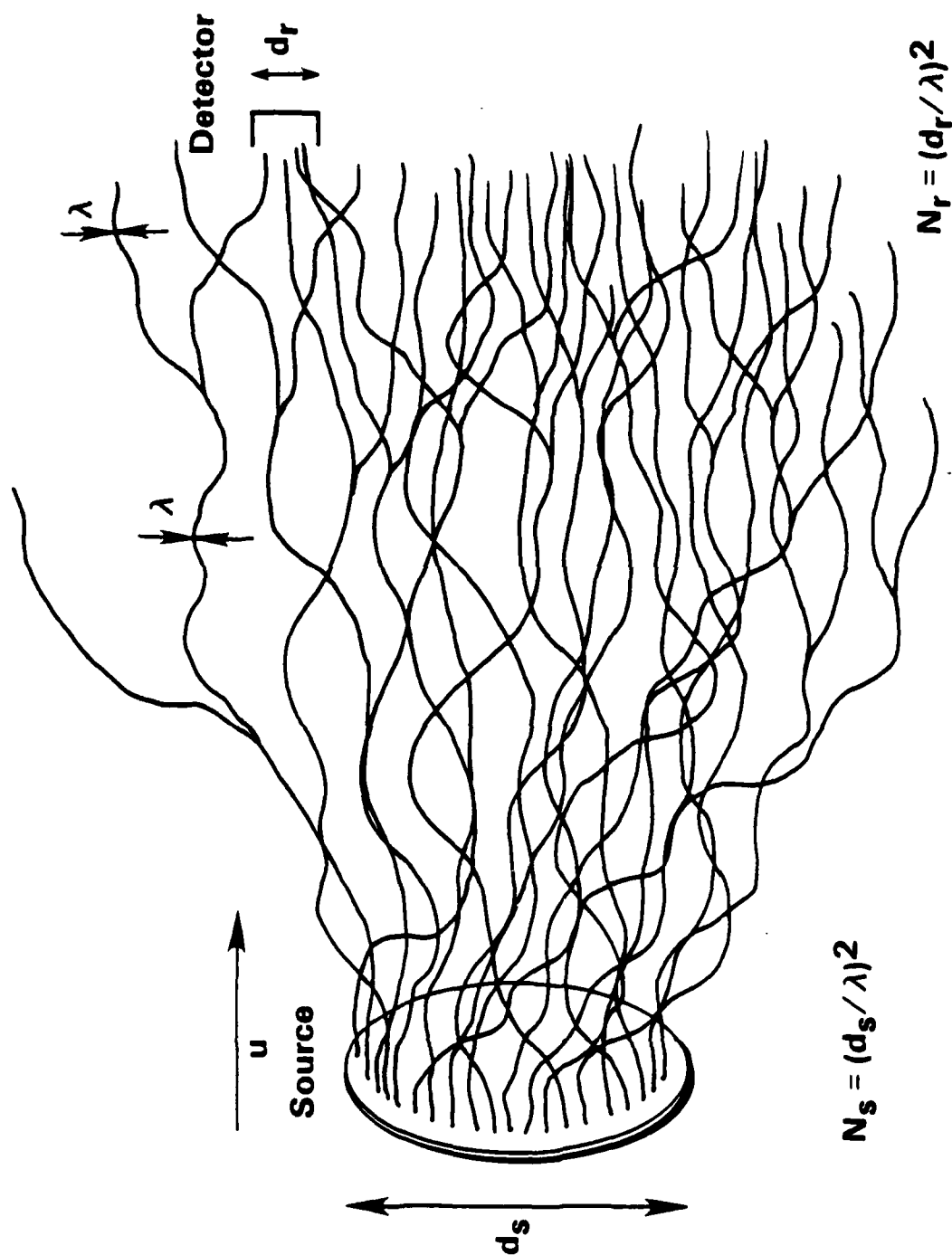
$$C(x,y,y) = C_o D(x,y,z) \tag{3}$$

Figure 1 Visualization of the Strand Concept. N_s Strands are Emitted and Subsequently Experience Dispersion among "Clean Air" Strands (not shown). Individual Strands Undergo Diffusive Growth and Split. A Receptor then Samples a Cluster of N_r Strands; a Fraction ρ of which Contain Pollution.

where the instantaneous plume dilution function $D(x,y,z)$ is defined such that at the source $D(x,y,z) = D(0,0,0) = 1$. The dilution function is assumed to be known, through means of a numerical or analytic model, but is given for the Gaussian plume model as

$$D(x,y,z) = \frac{\sigma_y(0)\ \sigma_z(0)}{\sigma_y(x)\ \sigma_z(x)}\ \exp\ [-\frac{1}{2}\ (y/\sigma_y(x))^2 - \frac{1}{2}\ (z/\sigma_z(x))^2]\ ,\quad (4)$$

where $\sigma_y(x)$ and $\sigma_z(x)$ are the horizontal and vertical instantaneous plume width standard deviations respectively as a function of downwind distance x. Substituting Eqs. (2b) and (4) into (3) gives us back a Gaussian plume expression and indicates that nothing unusual results from this re-grouping of terms.

Ignoring the minor "defect" introduced by the assumed Gaussian shape at the source, earlier assumed to be uniform, we note that when the dilution function takes on a maximal value of unity all available strands are populated with pollutant, as is consistent with our definition of the number of polluted strands N_s by Eq. (1). Temporarily ignoring diffusive breakup or splitting of the strands, these entities would simply disperse relative to each other, so that at some downwind receptor sampling strands of air, a fraction $D(x,y,z)$ would contain pollutant and a fraction 1-D would contain only "clean ambient air". Thus, we may think of $D(x,y,z)$ as either the usual diffusion function or as the polluted strand density function representing that fraction of locally available strands that are polluted. With this information in hand, one could immediately write down expressions for mean square concentration, concentration variance, and other moments of the distribution; however, the results would be in poor agreement with experiments as the all-important diffusive growth of the strands and subsequent strand breakup has been ignored.

If the strands experience diffusive forces specified by the
molecular diffusivity ν, they will double in areal size in a time,
t_d, given as

$$t_d = \lambda^2/(4\pi\nu) \tag{5a}$$

whereupon the strand is sheared apart by turbulent eddies into two
strands of diameter λ. Given molecular diffusivities of order
$10^{-5}m^2/sec$ and $\lambda \simeq 10^{-3}m$, one computes t_d in the $10^{-2} - 10^{-3}$
second range; however, as with λ, the precise numerical value of
t_d is not critical at this point except to note that it will be
sub-second and thus relatively short in comparison to many time scales
relevant to atmospheric dispersion. More generally we may merely say
that growth in the number of strands is associated with a time scale
τ without relating τ to t_d via a rigorous relation such as

$$\tau = t_d/\ln2 \qquad , \tag{5b}$$

that would result if we took the doubling concept literally. Allowing
such a growth in the number of polluted strands creates an interesting
dilemma that will prove quite useful in developing a differential
equation for the number density of polluted strands. Imagine that a
long pipe with diameter d_s were connected to the source described
earlier and that the flow speed u was such that the travel time in the
pipe was much greater than τ. N_s strands of concentration C_o
would go into the pipe and, since the pipe is completely filled with
strands, N_s strands of concentration C_o would come out the far
end. What if anything, happened during the journey along the pipe?
Diffusion and strand splitting did in fact occur, but since the pipe
was already as full as it could possibly be with a polluted strand
density of D = 1, the growing strands had no other choice but to
devour existing strands with the result that no net growth, g,
occurred in the polluted strand density. Can we then write down a
plausible differential equation describing the time evolution of the
effective polluted strand density, $\rho(t)$, defined as

$$\rho(t) \equiv g(t) \, D(t) \qquad , \tag{6}$$

where the dilution function $D(x,y,z)$ has been re-expressed as $D(t) = D(ut,y,z)$ for compactness? Knowing also that the number of strands will grow freely as $g \sim \exp(t/\tau)$ if the polluted strand density is low (i.e., $\rho \ll 1$) we may write the differential equation

$$\rho' = \rho(1-\rho)/\tau + g\ D' \quad , \tag{7a}$$

where the prime indicates the time derivative operator, as the simplest form that possesses the three necessary properties:

   a)   exponential growth of polluted strand fraction, ρ, with time scale τ for ρ small
   b)   termination of growth as ρ approaches unity via the term $(1-\rho)$, and
   c)   forced dilution created by changes in $D(t)$ in time.

Applying the chain rule to Eq. (6) and dividing through by D, Eq. (7a) may be alternatively expressed as

$$g' = g(1-\rho)/\tau \tag{7b}$$

which, because the factor $(1-\rho)$ corrects for new strands consuming or recombining with existing strands, elucidates the meaning of the term "net growth" function being applied to $g(t)$.

Equation (7a) for the polluted strand density or fraction is the principal result given to us by this simple, physically-based model of strand behavior, and this equation may be used in conjunction with any dispersion model, analytic or numerical, giving rise to a diffusion function D. It is also worth noting that since $\rho(t)$ is closely related to the intermittency factor γ discussed extensively by Wilson et. al. (1984), many useful expressions could be written down by inspection; however, we shall first explore the theory further.

2.2 Closed Form Solution

Without the forced dilution function D', Equation (7a) is identical to that used by population biologists to describe population

growth in a world with a fixed and finite food supply and is easily
solved analytically by means of the substitution $y = 1/\rho$. (Kaplan,
1958). Using this hint it quickly follows that <u>a solution</u>[+] to
Equation (7b) is

$$g(t) = \left\{ e^{-t/\tau} + \frac{1}{\tau} \int_0^t dt' \, e^{(t'-t)/\tau} D(t') \right\}^{-1} \tag{8}$$

where t' is simply a dummy variable for the time between release
(i.e., $t = 0$) and present time t. The only thing disturbing about
Eq. (8) is that the path between source and end point at time t is not
specified. Figure 1 shows how many different paths, equivalent to the
strand trajectories, could end up at a specific point and that some
weighted consideration of all possible paths (and thus strand density
time developments) should be involved in the correct solution of
Eq (7b). A plausible solution might consider the straight line path
as the mean solution pathway with the reciprocity theorem suggesting
that $D(t')$ in Eq. (8) be replaced by

$$D(t') = \frac{1}{2\pi \sigma_y (t-t') \sigma_z (t-t')} \int\int_{-\infty}^{+\infty} dy' dz' D(ut', y_m + y', z_m + z')$$

$$\cdot \exp\left\{ -\frac{1}{2} \frac{(y_m - y')^2}{\sigma_y^2 (t-t')} - \frac{1}{2} \frac{(z_m - z')^2}{\sigma_z^2 (t-t')} \right\} \tag{9}$$

where $y_m = yt'/t$ and $z_m = zt'/t$ specify the mean path and y', z'
are dummy integration variables. While Eq. (9) may represent a
significant theoretical point, it appears to be of secondary
importance in most applications. In addition, we note that Eq. (9)
can be cumbersome and numerically problematic to evaluate even for a
single mean path; thus, we turn to numerical integration of the more
well-conditioned Eq. (7b).

[+]Non-linear differential equations may have multiple solutions.

2.3 Numerical Solution

Numerical integration of Eq. (7b), subject to the initial
condition g(0)=1, must recognize that the potentially explosive growth
(i.e., exponential) in the solution could lead to rapid divergence of
the numerical solution from the true solution. For this reason we
adopt a second-order accurate, time-centered (i.e., Crank-Nicolson)
procedure to provide a reasonable compromise between accuracy and
stability. The finite difference form of (7b) then becomes

$$g_{n+1} = g_n + \beta\ (g_{n+1} + g_n)(1-\tfrac{1}{2}\ D_{n+1}g_{n+1} - \tfrac{1}{2}\ D_n g_n) \qquad (10)$$

where $\beta = \tfrac{1}{2}\ \Delta t/\tau$ and Δt is the time step. The time advanced
value, g_{n+1}, appears on both sides of the equation, and solution of
the resulting quadratic equation for g_{n+1} yields the familiar

$$g_{n+1} = \frac{-b + (b^2-4ac)^{1/2}}{2a} \qquad (11)$$

where $a = \tfrac{1}{2}\ \beta\ D_{n+1}$,

$b = 1 - \beta\ [1-\tfrac{1}{2}\ g_n(D_n + D_{n+1})]$,

and $c = -g_n\ [1 + \beta(1-\tfrac{1}{2}\ g_n D_n)]$.

Equation (11) is then marched in time using a time step chosen as the
lesser of 0.1τ and $0.1\ D/D'$.

Figure 2 shows several crosswind profiles of the resulting ρ
function at several values of downwind distance x=ut using parameters
relevant to the wind tunnel studies of Fackrell and Robins (1982).
The most interesting feature of these curves is the variation in
crosswind profile shape as one moves to different downwind distances.
Profiles with a fairly small plume-axis value of ρ, denoted ρ_0,
look Gaussian in shape while those with $\rho_0 \simeq 1$ show a broad
flat-top region with a precipitous drop at large y/σ_y. As it will
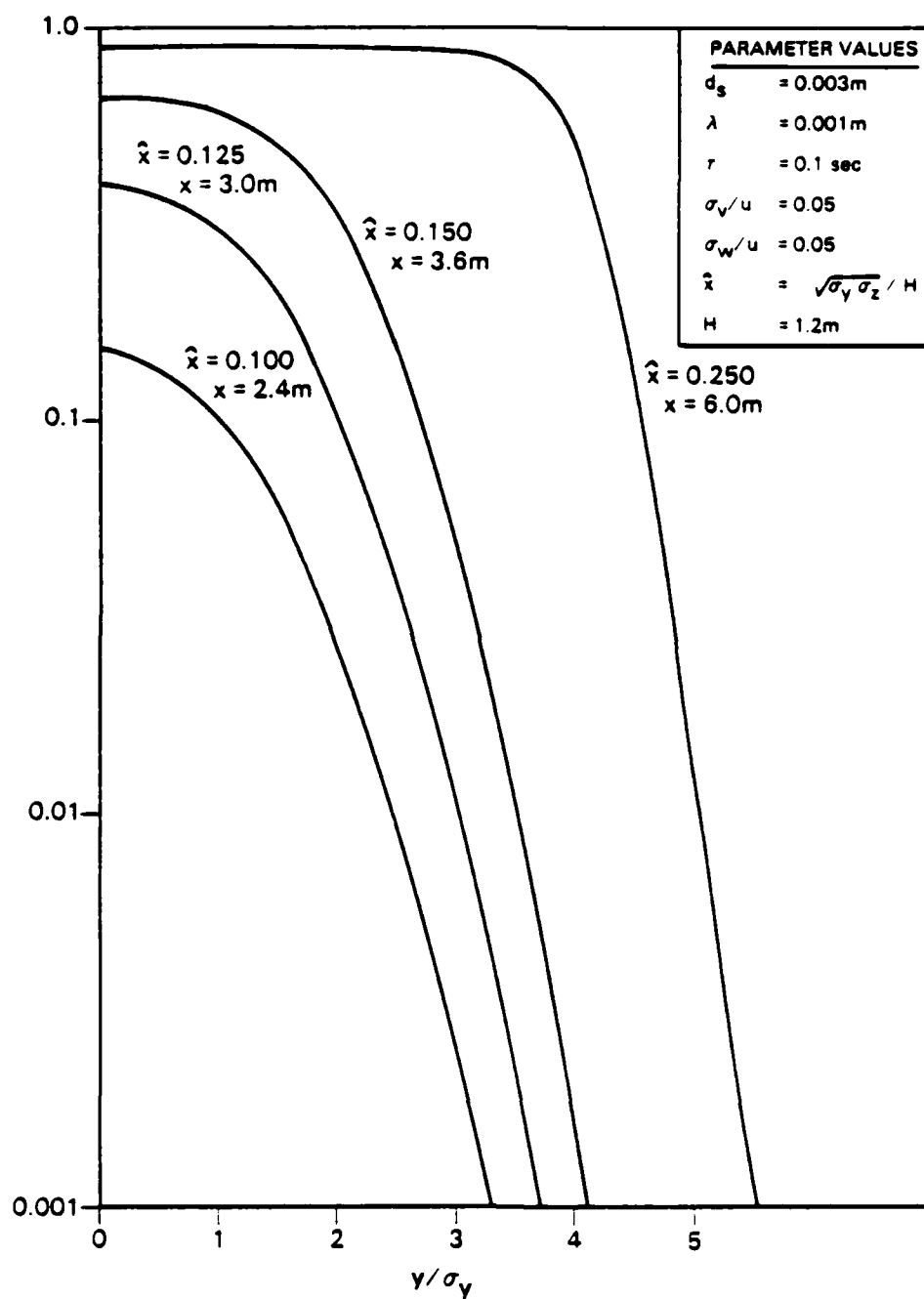prove important in the inclusion of plume meander effects to know this

Figure 2 Crosswind σ Profiles for Several Downwind Distances.
Distances are given in Meters and in Normalized Distance
Assuming a Boundary Layer Depth of 1.2m.

crosswind profile function, the crosswind behavior of the analytic
solution, Eq. (8), was considered assuming that the on-axis values
were available from the numerical integration procedure described
above. For the particular trajectory that follows the path
$y = \epsilon_y \sigma_y$, $z = \epsilon_z \sigma_z$, it can be shown that

$$\rho(ut,y,z)/\rho_0 = \frac{S(ut) \cdot \exp[-\frac{1}{2}(\epsilon_y^2 + \epsilon_z^2)]}{1 + \exp[-\frac{1}{2}(\epsilon_y^2 + \epsilon_z^2)][S(ut)-1]} \qquad (12)$$

where $\rho_0 = \rho(ut,0,0)$

and $S(ut) = e^{t/\tau}/g(t) = D(ut,o,o)e^{t/\tau}/\rho_0$. The function
$S(ut)$ is just the ratio of what g could have been (i.e., $e^{t/\tau}$) if
its growth were unhindered to what it actually is on the plume
centerline and hence it will be referred to as the "growth suppression
factor" S. Equation (12) provides a reasonable description of the
crosswind profiles shown in Figure 2 with the exception that falloff
is too rapid beyond when $\rho/\rho_0 \simeq 10^{-2}$, but this may not be a
serious drawback since this plume fringe regime should not dominate
most practical problems.

2.4 Statistical Measures

Once the mean probability ρ is known at the receptor, the
binomial distribution (Ross, 1972) gives the probability P(j), that j
of the N_r detected strands contain pollutant, as

$$P(j) = \frac{N_r!}{(N_r-j)!j!} \rho^j (1-\rho)^{N_r-j} \qquad j = 0,1,\ldots,N_r$$

where $N_r = (d_r/\lambda)^2$ for a receptor having an aperture d_r.
Thus, the probability distribution for detecting j polluted strands is
identical to those used for predicting the probabilities in die
($\rho = 1/6$) and coin ($\rho = 1/2$) toss games.

Since, there is always the finite probability

$$P(0) = (1-\rho)^{N_r}$$

that none of the strands contain pollutant, the "intermittency" factor

$$\gamma = 1-P(0), \qquad\qquad\qquad (15)$$

discussed extensively by Wilson et al. (1984), will be less than one even for an ideal, zero concentration threshold detector and in the complete absence of plume meander, as presently assumed here.

Figure 3 shows the intermittencies predicted by Eq. (15) for plausible strand properties of $\tau \simeq 0.2$ sec and $\lambda \simeq 0.4$ mm and superimposed onto wind tunnel measurements of Fackrell and Robins (1982). The curves, one for each of the five elevated source diameters considered, demonstrate that the strand theory approach can

i)   yield predictions that exhibit reasonable qualitative
     behavior in terms of source size and downwind distance
     dependence and

ii) span the range of observed values.

However, as important ingredients (e.g., plume meander, finite detector thresholds and averaging times, strand concentration distribution assumptions) are still missing from the formulation, results depicted should only be taken as grounds for "cautious optimism."

The moment generating function for the binomial distribution gives the ensemble average number of polluted strands sampled as
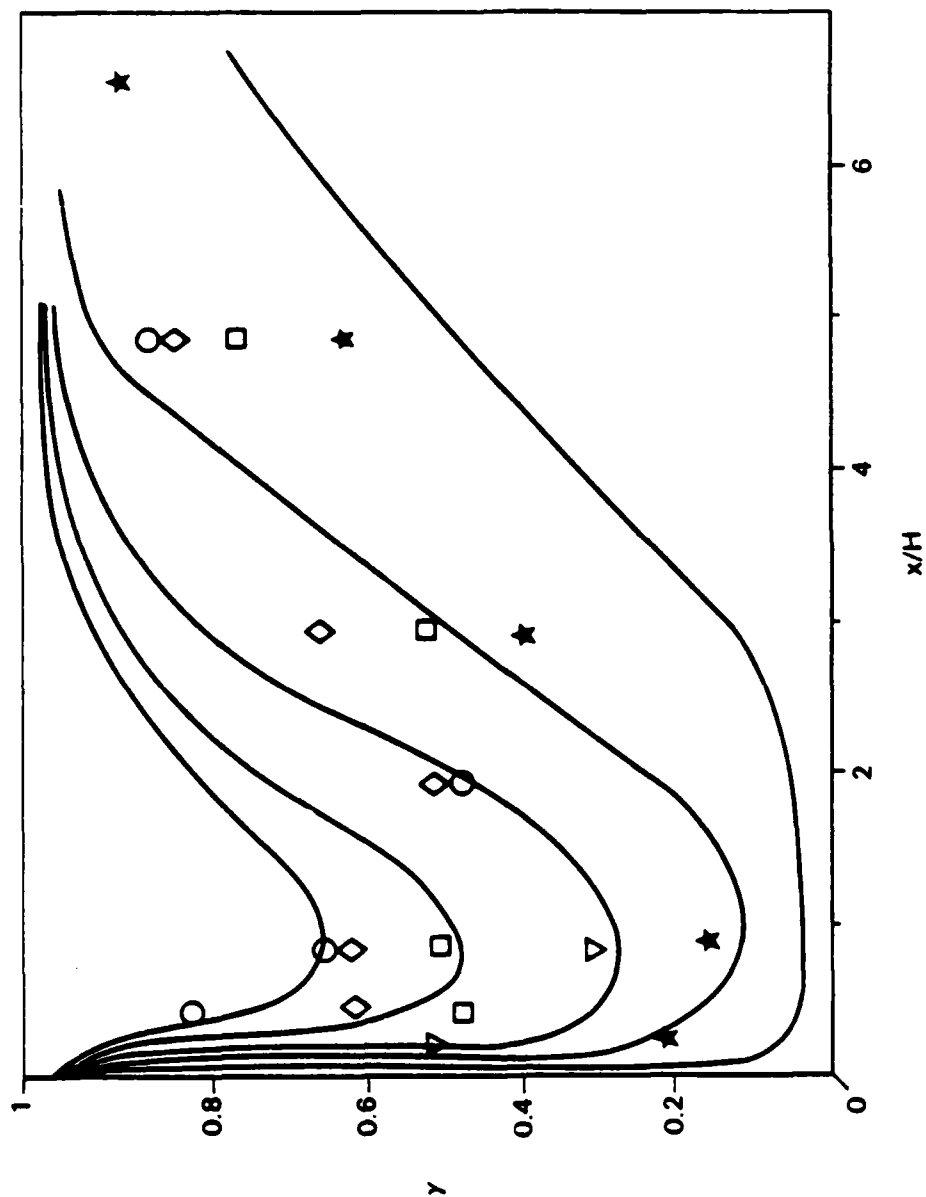
$$\bar{j} = N_r \rho \qquad , \qquad\qquad\qquad (16)$$

Figure 3   Plume-axis (x, o, z_s) Intermittency Factors Measured in the Fackrell and Robins (1982) Wind Tunnel Study. Solid Curves are Eq. (15) Predictions for the Five Elevated Source Diameters of 0.30, 0.85, 1.50, 2.50 and 3.50 Centimeters. (The lowest curve corresponds to the smallest source).

the ensemble mean square number of polluted strands as

$$\overline{j^2} = N_r(N_r-1)\rho^2 + N_r\rho \qquad , \qquad (17)$$

as well as any higher order measure $\overline{j^n}$. Thus, the ensemble variance in the number of polluted strands is just

$$\sigma_j^2 = \overline{j^2} - \overline{j}^2 = N_r\rho(1-\rho) \qquad , \qquad (18)$$

so that, even if at each point all strands have the same concentration, a finite ensemble concentration variance would be observed.

Hypothesizing that at the receptor each of the j polluted strands has the same concentration, C_s, the particular sample will have concentration

$$C(j) = C_s(j/N_r) \qquad (19)$$

and the ensemble mean concentration will be

$$\overline{C} = \sum_j P(j)C(j) = C_s(\overline{j}/N_r) = C_s\rho \qquad (20)$$

However, for this result to be consistent with Eq. (3) it must be true that

$$C_s = C_o/g(t) \qquad . \qquad (21)$$

Similarly, the squared concentration

$$c^2(j) = [C_s(j/N_r)]^2 = j^2[C_o/(gN_r)]^2 \tag{22}$$

results in the ensemble mean square concentration

$$\overline{c^2} = \sum_j P(j)c^2(j) = [(1-1/N_r)\rho^2 + \rho/N_r](C_o/g)^2 \tag{23}$$

and variance of

$$\sigma_C^2 = \overline{c^2} - \overline{c}^2 = \frac{1}{N_r}\rho(1-\rho)(C_o/g)^2 \tag{24}$$

The total concentrations fluctuation intensity, i, can then be expressed as

$$i^2 = \sigma_C^2/\overline{c}^2 = \frac{1}{N_r}(1/\rho - 1) \tag{25}$$

Wilson et al. (1984) discuss the logic and convenience of decomposing the fluctuation intensity, i, into two parts: the part arising from detecting positive concentrations and the part due to samples where the detector sees no material. The resulting decomposition is

$$i^2 = i^2_p/\gamma + (1-\gamma)/\gamma \quad , \tag{26}$$

where

$$i_p^2 = \sigma_{Cp}^2/\overline{C}_p^2 \quad , \tag{27}$$

and the subscript p refers to that fraction γ of the samples where the receptor conditionally sees positive (i.e., finite non-zero) signals. This decomposition into conditional quantities, denoted by the subscript p, is also convenient for consideration of the effects of allowing strands to have a distribution of concentrations.

2.5 The Exponential Strand Concentration Distribution

In the previous section, all strands at a given point in space were assumed to have the same concentration C_s; however, a receptor at some downwind location will detect strands that have experienced a spectrum of histories as opposed to the simplistic notion that each strand will have suffered g(t)-1 bifurcations. In developing a theory of strand "ageing" as a sequence of bifurcations, analogy (Ross, 1972) with other decay distribution examples (e.g., radioactive decay, probabilistic failure analysis) suggests that the strands will have an exponential age distribution

$$P(r) = \frac{1}{\alpha} e^{-r/\alpha} \tag{28}$$

where r is the age parameter and α represents the average strand age. As increasing strand age corresponds to a greater number of bifurcations, yielding successively lower strand concentrations, it is reasonable to conjecture that these concentrations also will be distributed exponentially as

$$C_s(r) = C_o e^{-r/\delta} \tag{29}$$

where δ is a constant [+] describing the effect on concentration of a single bifurcation. The average strand concentration, which Eq. (21) tells us must be $C_o/g(t)$, is then

———————

[+] A value of δ = ln2 would yield $C_s(r) = C_o(\frac{1}{2})^r$ implying that each effective bifurcation reduced the strand concentration in half, but such a strong assumption is not necessary.

$$\bar{C}_s = \frac{C_0}{\alpha} \int_0^\infty dr \, \exp[-r(1/\alpha + 1/\delta)] \qquad (30)$$

$$= C_0/(1 + \alpha/\delta) = C_0/g(t)$$

which therefore constrains the ratio α/δ to be

$$(\alpha/\delta) = g(t) - 1 \qquad (31)$$

and leads to the same ensemble mean concentrations of

$$\bar{C} = C_0 \rho/g \qquad (32)$$

obtained assuming uniform concentration strands. Perhaps, not surprisingly, the average age parameter α has a time dependence proportional to $g-1$. At the source $g=1$ and the distribution becomes an infinite spike at $r=0$ (i.e., the delta function $\delta(r)$) corresponding to the emission of unaged strands of concentration C_0. It is also noteworthy that, even at great distances from the source, there will be a finite probability of detecting an undiluted strand with the original concentration C_0.

Computation of higher moments of the observed concentration must recognize that for any one sample only j of the N_r strands are populated with pollutant. Thus, the mean square concentration for j polluted strands involves the j-dimensional integral

$$\overline{C^2}(j) = \frac{C_0^2}{N_r^2 \alpha^j} \int_0^\infty dr_1 \int_0^\infty dr_2 \cdots \int_0^\infty dr_j [e^{-r_1/\delta} + e^{-r_2/\delta} + \ldots + e^{-r_j/\delta}]^2$$

$$\cdot \, e^{-(r_1 + r_2 + \ldots + r_j)/\alpha} \qquad (33)$$

$$= \frac{C_0^2}{N_r^2} \left[\frac{j}{1 + 2\,\alpha/\delta} + \frac{j^2 - j}{(1 + \alpha/\delta)^2} \right] \quad ,$$

and the ensemble mean square concentration

$$\overline{c^2} = \sum_j P(j)\overline{c^2(j)}$$

involving the sum over j from zero to N_r becomes

$$\overline{c^2} = \frac{c_o^2}{N_r^2} [\frac{N_r\rho}{1+2\alpha/\delta} + \frac{N_r(N_r-1)\rho^2+N_r\rho-N_r\rho}{(1+\alpha/\delta)^2}]$$

after invoking the expressions for mean j and j^2 given by Eqs. (16) and (17). Cancelling $N_r\rho$ contributions in the second term and replacing α/δ with g-1 then yields the final results

$$\overline{c^2} = \frac{c_o^2}{N_r}[\frac{\rho}{2g-1} + \frac{(N_r-1)\rho^2}{g^2}] \quad \text{and} \tag{34}$$

$$\sigma_c^2 = \frac{c_o^2}{N_r}[\frac{\rho}{2g-1} - (\rho/g)^2] \tag{35}$$

for the ensemble mean square concentration and concentration variance respectively. The corresponding conditional estimates can then be immediately written down as γ^{-1} times the corresponding ensemble mean values given by Eqs. (32) and (34), since excluding the j = 0 terms from the sum requires renormalization of the probabilities P(j) by the factor $\gamma^{-1} = (1-P(0))^{-1}$ to ensure that the conditional probabilities $P_c(j)$ obey the constraint

$$\sum_j P_c(j) = 1 \qquad j = 1,2,\ldots N_r \qquad ,$$

where $P_c(j) = P(j)/\gamma$. The equivalent conditional variance is then just

$$\sigma_{cp}^2 = \frac{C_o^2}{\gamma N_r} \left[\frac{\rho}{2g-1} - (\rho/g)^2 (N_r(1/\gamma-1)+1)\right] \qquad (36)$$

and the conditional concentration fluctuation intensity, i_p, given by

$$i_p^2 = \frac{\gamma}{N_r}\left[\frac{g}{D(2g-1)} - N_r(1/\gamma-1)-1\right] \qquad . \qquad (37)$$

Far from the source $g \gg 1$ and $D \ll 1$ so that the first term dominates and $i_p \simeq [\gamma/(2N_r D)]^{1/2}$, revealing the increasing fluctuation intensity as one moves away from the plume centerline.

3. Inclusion of Additional Effects

The theory development to this point has focussed on the problem of determining ensemble statistical measures. Experimental evaluation of these measures would require repeated sampling at fixed points relative to the centerline of the instantaneous plume. While perhaps not an impossible experimental feat, it is of greater relevance to adapt the theory to the case of a plume meandering back and forth across a receptor fixed in space and accepting samples without regard to the plume's position.

In the case of the Gaussian plume model example we are able to build upon the meandering plume formalism developed by Gifford (1959). Defining a coordinate system where the receptor is located at a point (y,z) relative to the mean centerline position of the plume, the position of the receptor relative to the plume centerline at any instant is $(y-y',z-z')$ and occurs with probability

$$P(y',z') = \frac{1}{2\pi\sigma_{yM}(x)\sigma_{zM}(x)} \exp\left[-\frac{1}{2}(y'/\sigma_{yM}(x))^2 - \frac{1}{2}(z'/\sigma_{zM}(x))^2\right] \qquad ,$$

neglecting the influence of ground reflections. Referring back to
Eq. (32) and recalling that $D = \rho/g$ we may immediately recompute the
mean concentration as

$$\bar{C} = C_o \int\int_{-\infty}^{+\infty} dy' dz' P(y',z') D(x,y-y',z-z') = C_o D_T(x,y,z) \qquad (39)$$

where D is given by Eq. (4) and D_T has the same form as D but uses
the total sigma, $\sigma_T = (\sigma^2 + \sigma_M^2)^{1/2}$ in place of the
instantaneous sigma measures σ, referred to as σ_y and σ_z
throughout this paper.

Computation of the mean square concentration involves a similar
integral averaging of Eq. (34) and can be written

$$\overline{C^2} = \frac{C_o^2}{N_r} \int\int_{-\infty}^{+\infty} dy' dz' P(y',z') [\frac{\rho}{2g-1} + (N_r-1)D^2] \qquad (40)$$

where

$$D = D(x,y-y',z-z') \quad ,$$
$$\rho = \rho(x,y-y',z-z') \quad ,$$
$$g = \rho/D$$

and $\rho(x,y-y',z-z')$ can be approximated with the help of Eq. (12).
Given the complexity of Eq. (12), evaluation of Eq. (40) will either
require additional approximations or numerical integration.

Inclusion of the effects of a finite detector concentration
threshold, C_T, is another complexity that may require numerical
evaluation in most cases. Referring back to Eq. (33), the generalized
form of the nth moment may be expressed as

$$\overline{C^n(j)} = (C_o/N_r)^n \alpha^{-j} \int_o^\infty dr_1 \int_o^\infty dr_2 \cdots \int_o^\infty dr_j [e^{-r_1/\delta} + e^{-r_2/\delta} + \ldots + e^{-r_j/\delta}]$$

(41)

$$\cdot e^{-(r_1+r_2+\ldots r_j)/\alpha} \theta(e^{-r_1/\delta} + e^{-r_2/\delta} + \ldots + e^{-r_j/\delta} - C_T N_r/C_o)$$

where the theta function $\theta(x)$ is defined as

$$\theta(x) = \begin{cases} 0 \text{ for } x < 0 \\ 1 \text{ for } x \geq 0 \end{cases}$$

Need for the θ-function disappears as $C_T \rightarrow 0$ and can be replaced by altered integration limits for $N_r = 1$ or 2, but we have not yet developed a general, analytic solution for Eq. (41); however, integrals of this type are relatively common in statistical mechanics. Once accomplished, one could compute the weighted sum over j followed by the integral averaging suggested by Eq. (40).

Another important effect of the θ function in Eq. (41) is that the $n = 0$ normalization moment is less than unity so that the probabilities given by Eq. (13) are modified and become

$$P'(j) = P(j) \overline{C_o(j)}$$

(42)

This has the immediate effect of reducing the value of γ from that given by Eq. (15) to

$$\gamma = 1 - P(0) - \sum_{j=1}^{N_r} P(j)[1-\overline{C^o(j)}]$$

(43)

thus, indicating the significance of instrumental "details" on the outcome of concentration fluctuation experiments.

Perhaps the last refinement that would be considered essential for application to experimental data is the correction for averaging time. For very small averaging times, τ_a, much smaller than the Lagrangian time scale, τ_L, associated with the principal, meander driving eddies, it is thought that the temporal smearing is equivalent to an increased receptor aperture diameter d_r' defined as

$$d_r' = d_r + (\sigma_v \sigma_w)^{1/2} \tau_a \quad , \qquad (44)$$

where σ_v and σ_w are the relevant lateral and vertical turbulent velocities. This then has the effect of increasing the number of strands received by the receptor to

$$N_r' = (d_r'/\lambda)^2 = N_r [1 + (\sigma_v \sigma_w)^{1/2} \tau_a / d_r]^2 \qquad (45)$$

which in turn has the effect of lowering the magnitude of the concentration fluctuations as given by Eq. (35) for example.

Extension of the theory to longer time averaging periods remains to be worked out in detail, but may require further assumptions about the form of the auto-correlation function and τ_a/τ_L. Venkatram (1979) provides results of assuming an exponential form for this function and Hanna (1984) discusses other treatments of this problem.

Finally, it should be noted that the strand theory can easily be extended to multiple species. This extension could be an important step in modeling the influence of sub-grid scale inhomogeneities on processes involving non-linear chemical reactions.

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